

CDG-101US

PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Appl. No: 10/051,995
Applicant: Thomas E. McWhorter et al.
Filed: January 18, 2002
Title: METHOD AND APPARATUS FOR GENERATING GASEOUS
CHLORINE DIOXIDE-CHLORINE MIXTURES
TC/A.U.: 1754
Examiner: Ngoc Yen M. Nguyen
Confirmation No.: 9137
Docket No.: CDG-101US

DECLARATION UNDER 37 C.F.R. §1.131
OF JAMES C. SIMMONS

Honorable Commissioner of Patents and Trademarks
Alexandria, VA 22313-1450

S I R :

I, James C. Simmons, do hereby declare and state the following.

I am the attorney of record for above-identified patent application.

I declare that the inventions as described by claims 1, 3-38 of the subject patent application were conceived and reduced to practice in the United States prior to January 25, 2001.

Exhibits A1-A10 are true copies of pages of a laboratory notebook kept in the regular course of business by the inventor/applicants as employees and principal of the assignee of the above identified application. The assignment is recorded in the records of the United States Patent and Trademark Office at Reel 012519 Frame 0703.

That I have reviewed the notebook and procedures of the assignee to verify the notebook was a record kept in the normal course of business by Applicants' assignee.

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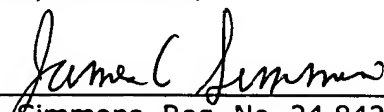
Exhibits A1-A10 show the results of various tests run by or at the direction of Applicants wherein Sodium Chlorate was reacted with Hydrochloric Acid to produce a mixture of Chlorine Dioxide and Chlorine as claimed in the application.

Dates on Exhibits A1-A10 have been replaced by a date range "Before June 25, 2001".

In view of the fact that the invention of Applicants, as disclosed and claimed in the above-identified application, was conceived and actually reduced to practice prior to June 25, 2001, U.S. Patent Publication 2003/0007899 is not a prior art reference against the invention of claims 1, 3-38 of the above-identified application.

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and, further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Respectfully submitted,



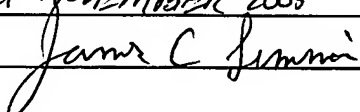
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The Commissioner for Patents is hereby authorized to charge payment to Deposit Account No. 18-0350 of any fees associated with this communication.

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21 NOVEMBER 2005


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EXPERIMENT 11

BEFORE JUNE 25, 2001

3

NaClO_3 (400 g/l) changed at 500 ml.

HCl (15% w/w) changed at 300 ml.

20 min just before after not connected to reactor.

10 min just before flow connected to reactor.

30 min Time first injection of 50 ml acid with start-up of sparging.

Temperature @ 80°C in reactor with sparge

1 hr Time. Second injection of 50 ml acid with sparging.

Water flow rate through the house is 2000 l/min. It is measured at the time of daily operation.

90 min Time. Start of the 3rd injection of 50 ml acid with sparging.

2 hr Time. 4th injection of 50 ml acid with sparging.

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Before June 25, 2011

EXHIBIT A-2

This run is related under Group #20.

Sample #3.

At 150 min Time, 5th injection of 50 ml acid with sparging.

At 3 min Time. The 6th injection of 50 ml acid with sparging.

At 225 min Time. Turned off the heater of reactor bath and took the reactor out of the bath. Sparging is continued in 5 minutes. Temperature of reactor dropped to 74°C.

At 235 min Time. Stopped the sparging.

The Data are stated in File #4 in Hatch and in C:\Boragin\Experiment\20010329.xls

The final volume of liquid in reactor of and is 625 ml.

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BEFORE JUNE 25, 2001

EXHIBIT A3

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NaClO_3 changed of 500 mL.

HCl changed of 400 mL.

No Sparging is applied throughout.

• 0 min Time, 1st injection of 80 mL acid.

• 5 min Time, 2nd injection of 80 mL acid.

• 55 min Time, 3rd injection of 80 mL acid.

• 90 min Time, 4th injection of 80 mL acid.

• 130 min Time, 5th, last injection of 80 mL acid.

• 170 min Time, took the reactor out of

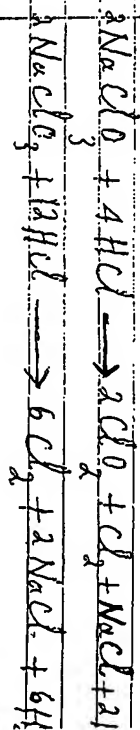
the water bath and started sparging. This

is confirmed with the reactor temperature

drops down. Time is of 2:50 at start of

this step.

Sample	A	B	C	D	thrust
injection	mL	mL	mL	mL	typ
1	50	3.54	4.91	0.10	0.00
2	50	4.95	7.90	2.9	0.20
3	50	4.85	8.44	3.23	0.13
4	50	3.76	13.81	3.62	1.45
5	50				



V₁ I₁ K₁ A₁ D₁ C₁ S₁

V₂ I₂ K₂ A₂ D₂ C₂ S₂

V₃ I₃ K₃ A₃ D₃ C₃ S₃

V₄ I₄ K₄ A₄ D₄ C₄ S₄

V₅ I₅ K₅ A₅ D₅ C₅ S₅

V₆ I₆ K₆ A₆ D₆ C₆ S₆

V₇ I₇ K₇ A₇ D₇ C₇ S₇

V₈ I₈ K₈ A₈ D₈ C₈ S₈

V₉ I₉ K₉ A₉ D₉ C₉ S₉

V₁₀ I₁₀ K₁₀ A₁₀ D₁₀ C₁₀ S₁₀

V₁₁ I₁₁ K₁₁ A₁₁ D₁₁ C₁₁ S₁₁

V₁₂ I₁₂ K₁₂ A₁₂ D₁₂ C₁₂ S₁₂

V₁₃ I₁₃ K₁₃ A₁₃ D₁₃ C₁₃ S₁₃

V₁₄ I₁₄ K₁₄ A₁₄ D₁₄ C₁₄ S₁₄

V₁₅ I₁₅ K₁₅ A₁₅ D₁₅ C₁₅ S₁₅

V₁₆ I₁₆ K₁₆ A₁₆ D₁₆ C₁₆ S₁₆

V₁₇ I₁₇ K₁₇ A₁₇ D₁₇ C₁₇ S₁₇

V₁₈ I₁₈ K₁₈ A₁₈ D₁₈ C₁₈ S₁₈

V₁₉ I₁₉ K₁₉ A₁₉ D₁₉ C₁₉ S₁₉

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Water stock acid EXHIBIT A4
BEFORE JUNE 25, 2001

300 450

150

NaClO₃ changed at 250 mL (400 g/L)

HCl changed at 480 mL (20% W/W)

Water flow rate is 130 mL/sec.

At 5' 1st injection of 80 mL acid.

No spraying is used throughout.

At 40' 2nd injection of 80 mL acid.

At 75' 3rd injection of 80 mL acid.

At 120' 4th injection of 80 mL acid.

$$\text{mg/L Cl}_2 = \frac{\text{Absorbance}}{0.0204}$$

Water flow rate = 7.8 L/min.

At 170' 5th injection of 80 mL acid.

At 210' 6th, last injection of 80 mL acid.

Started to collect Cl₂ sample at 210' Time
stopped to collect Cl₂ sample at 240' Time.

Hack stopped recording at 4:00 Time.

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BEFORE June 25, 2001

Data on steel to fill # 8, April 3. Sample 2
group 20.

At 170' 4:15 Time started spraying and to

the reaction flask out of water bath.

At 170' 4:47 Time. Disconnected ejector from

reaction flask at 50'C.

Data was started to fill # 9.

Measurement of Chlorine in the collected sample

from injection # 6.

mL sample	A	B	C	D	Reagent	mg/L
10	1.14	1.32	0.55	0.00	PAC	16.2

Total volume of Cl₂ sample:

$$30 \text{ min} \times 8 \text{ L/min} = 240 \text{ L}$$

$$\# \text{ moles Cl}_2 = 16.2 \text{ mg/L} \times 240 \text{ L} \times \frac{1}{1000} = 0.05 \text{ moles}$$

General Results of the Experiment:

Total Acid changed # moles HCl:

$$480 \text{ mL} \times 1.1 \frac{\text{g}}{\text{mL}} \times \frac{20 \text{ g HCl}}{100 \text{ g solution}} = 36.5 \frac{\text{g}}{\text{mole}}$$

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BEFORE JUNE 25, 2001

EXHIBIT 45

BEFORE JUNE 25, 2001

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Total ClO_2 produced # moles = 0.61 moles ClO_2 Total NaClO_3 used in Reaction 1 = 0.61 molesAcid used in Reaction 1 = 1.2 moles HCl

Acid used in Reaction 2 = 2.9 - 1.2 = 1.7 moles

Total NaClO_3 used in Reaction 2 = $\frac{1.7}{6} = 0.28$ Total NaClO_3 changed # moles = $2.50 \text{ mL} \times \frac{400 \text{ g}}{1000 \text{ mL}} / 106.4 \frac{\text{g}}{\text{mole}} = 0.94 \text{ moles}$ Total NaClO_3 left at end of experiment = $0.94 - (0.61 + 0.28) = 0.05 \text{ moles}$ Chlorine produced in Reaction 1 = $\frac{0.61}{3} = 0.3 \text{ moles}$ Chlorine produced in Reaction 2 = $3(0.28 + 0.05)$

= 1.0 moles

Total Cl_2 produced = 1.0 + 0.3 = 1.3 moles

In the next page the summary and the

real time data of the experiment are

presented. Temperature is 80°C throughout.

Total ClO_2 mg	41315
Total ClO_2 g	41.315
Total ClO_2 mole	0.6125
Total NaClO_3 g	100.00
Total NaClO_3 mole	0.8095
Yield %	66.20

Stage # 1: Fraction ClO_2	0.17
Stage # 2: Fraction ClO_2	0.24
Stage # 3: Fraction ClO_2	0.2
Stage # 4: Fraction ClO_2	0.17
Stage # 5: Fraction ClO_2	0.12
Stage # 6: Fraction ClO_2	0.09
Stage # 5: Fraction ClO_2	0.01
Total Fraction ClO_2	1.00

The above summary is based on real time data of the experiment. The main steps in manipulation of data are:

* Change Absorbance # to mg/L ClO_2

* Change mg/L to mg/min ClO_2

* Integrating point data using Simpson's

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EXHIBIT #6

11

BEFORE JUNE 25, 2001

Cl₂ mg/L
14.0

Cl₂ mg/L
15.2

The velly last pack is got sparging while the flask is taken out of water bath.

The average Hach reading for Cl₂ mg/L during injection #6 is 22.4 compared to

17.0 which is obtained above by titration

Reaction temperature was 80°C.

Total Cl₂ produced mg = 40,000 mg

very close to what obtained in experiment

20010403.

Cl₂ produced in injection #6 has an

average value of 19.0 mg/L which is

about 0.28 mol/L.

Cl₂ produced in injection #6 has an

average concentration of 15.2 mg/L which

is about 0.21 mol/L.

From coefficients in reactions 1 and 2

from reaction 1 and only 0.07 mol/L Cl₂ is produced from reaction 2. This means the reaction 1 happens 12 times faster than reaction 2 leading to an efficiency of 12/13 = 0.92 of injection #6.

In injection #6:

2.900 mg Cl₂ ≈ 0.043 moles Cl₂

HCl changed: $40.0 \times 1.1 \times 0.2 / 36.5 \approx 0.241$ moles

0.086 moles HCl for reaction 1

0.032 - Total moles Cl₂ produced

0.011 moles Cl₂ from reaction 2

0.022 + 0.086 = 0.108 moles HCl consumed in both reactions 1 and 2.

The final volume of liquid in the reaction flask was 55.0 mL.

moles HCl left unreacted in injection #6 is 0.241 - 0.108 = 0.133 moles HCl unreacted.

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EXHIBIT A7

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13

NaClO₂ changed at 250 ml (400 g/L).

HCl is changed at 480 ml (20% W/W).

Total number of injections will be #12.

Each injection is 48 ml of acid at an interval of every 15 to 20 minutes as marked on the next data list.

A	B	C	D	Sample ml	mg/L ClO ₂
0.45	2.34	0.35	0.08	10	22

The above list is for a Hach Sample 20350 absorbance that is 36.7 mg/L according to the calibration curve with formula:

$$\text{Absorbance} = 0.0204 (\text{ClO}_2 \text{ mg/L})$$

Cl₂ Sampling:

Started to collect for Cl₂ test at 6th injection stopped to collect for Cl₂ test at 11th injection after 15 minutes.

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EXHIBIT A-8

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NaClO₃ changed at 250 ml (400 g/L)

HCl changed at 480 ml (20% w/v)

Temperature is changed to 90°C throughout

Number of injections is #12, with 15 min between injections. The date the stored in full

The final liquid volume was 400 ml.

The yield is exactly the same as that obtained in list at 80°C. That is 40,000 mg ClO₂Te

18.8 g → 58.2 g

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EXAM 18 A9

BEFORE JUNE 25-2001

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1st: 80 mL	10 min.
2nd: 40 mL	10 min.
3rd: 40 mL	10 min.
4th: 80 mL	10 min.
5th: 80 mL	10 min.
6th: 80 mL	5 min.
7th: 80 mL	5 min.
Sparks	30 min.

NaClO₃ changed at 2.50 mL (4.00 g/L) ^{3.50 g/L}

HCl changed at 4.80 mL (2.87 g/L) ^{3.75}

Injections are done according to the chart.

A	B	C	D	chg	cl
mL	mL	mL	mL	mg/L	m
9.76	13.00	0.93	0.00	52.14	
Sample 2.5 mL	2.5	10	10		

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EXHIBIT A10

BEFORE JUNE 25, 2004

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Density of NaClO_3 solution:

Empty cylinder = 216 g.

250 mL in cylinder = 526 g.

Density = $\frac{526 - 216}{250} = 1.24 \text{ g/mL}$

The density corresponds to

about 450 g/L NaClO_3 .

1 80 10

2 40 10 250 mL NaClO_3 (450 g/L)3 40 10 480 mL HCl (20% w/v).

4 80 10

5 80 10

6 80 5

7 80 15

Liquor of 600 mL:

A B C D E

175 0.00 0.00 0.00 148

Sample 5 5 5 5 15*

* Diluted 20 times.

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